

UCRL-92728
PREPRINT

CIRCULATION COPY
SUBJECT TO RECALL
TWO WEEKS

A SEARCH FOR SUPERHEAVY ELEMENTS USING
THE $^{48}\text{Ca} + ^{254}\text{Es}$ REACTION

R. W. Lougheed
J. H. Landrum
E. K. Hulet
J. F. Wild
R. J. Dougan
A. D. Dougan
H. Gaggeler
M. Schadel
K. J. Moody
K. E. Gregorich
G. T. Seaborg



This paper was prepared for submittal to
Physical Review C
May 1985

Lawrence
Livermore
National
Laboratory

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

A search for superheavy elements using the $^{48}\text{Ca} + ^{254}\text{Es}$ reaction

**R.W. Lougheed, J.H. Landrum, E.K. Hulet, J.F. Wild,
R.J. Dougan, and A.D. Dougan
University of California, Lawrence Livermore National Laboratory,
Livermore, California 94550**

**H. G \ddot{a} ggeler and M. Sch \ddot{a} del
Gesellschaft f \ddot{u} r Schwerionenforschung, Darmstadt,
Federal Republic of Germany**

**K.J. Moody, K.E. Gregorich, and G.T. Seaborg
Lawrence Berkeley Laboratory, University of California,
Berkeley, California 94720**

ABSTRACT

We measured upper limits for the production of superheavy elements (SHEs) from the compound nucleus reaction $^{48}\text{Ca} + ^{254}\text{Es}$. This combination permits the closest approach to the predicted island of stability at the 184 closed-neutron shell of any practical fusion reaction. We used aqueous and gas-phase separations to isolate SHE fractions within an hour from the end of the bombardments. In these experiments we did not observe SHEs above a limit of $3 \times 10^{-31} \text{ cm}^2$ for spontaneous-fission half-lives from fractions of a day to a few months.

PACS Number: 25.70.Jj

Theoretical calculations by Randrup et al.¹ and Fiset and Nix² predict the maximum stability for the superheavy elements (SHEs) against spontaneous fission (SF) to be at the 184-neutron closed shell. However, SF half-lives differ by 10^9 between these predictions. The calculations of Randrup et al. show a more rapid decrease in fission barrier height below 184 neutrons, becoming only 3–4 MeV at 178 neutrons with corresponding SF half-lives of 10^{-16} years, as compared with years or more at 184 neutrons. The half-lives at or near 184 neutrons are thus dominated by alpha or beta decay.

This sensitivity of SF half-life to neutron number for the SHEs dictates a very small choice of nuclear reactions that can produce a product as close as possible to the 184-neutron shell. The much favored and extensively studied $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction would produce element 116 with 178 neutrons after emission of two neutrons from the compound nucleus. Upper limits for the production cross section of 10^{-34} to 10^{-35} cm^2 were measured for SHEs with half-lives as short as 10^{-6} s produced by this reaction.³ These low limits indicate either a half-life shorter by 10^{15} than the 130 y predicted by Fiset and Nix or a drastically lower production cross section. If the predictions of Randrup et al. are realistic, this nuclide would have a SF half-life of about 10^{-9} s and a fission barrier of only 4 MeV. A fission barrier below the neutron binding energy could result in a reduction of more than 10^9 in the reaction cross section due to fission during the de-excitation of the compound nucleus. The consequences of such reductions in yield or half-life make it imperative to choose a reaction that lands as close as possible to the 184-neutron shell, where the fission barriers are predicted to be highest. From this standpoint, the $^{48}\text{Ca} + ^{254}\text{Es}$ reaction is the most favorable compound-nucleus reaction currently available. For example, Randrup et al. predict fission barriers of greater than 6 MeV for the 2n-out product of the complete fusion reaction with Es and SF half-lives comparable to the alpha half-lives. A large additional gain in half-life should be made because the 2n out product from this reaction is an odd-odd nucleus. Bjørnholm

and Swiatecki noted the merits of the Es reaction in a detailed comparison of several reactions, including the ^{48}Ca reaction with ^{248}Cm .⁴

The closer approach to the 184-neutron shell appears to be a distinct advantage for the Es reaction vs. the Cm reaction. However, Leander et al. and Möller predict $Z = 114$ to be the most stable atomic number and the most stable neutron numbers to be nearer 176 to 178, where the shell correction is 8 to 9 MeV vs. 6 to 7 MeV at 184 neutrons.⁵ The Ca plus Cm reaction would then be the better reaction, because it would produce an isotope within the most stable region with 8-MeV shell correction energy. A single α -decay would lead to the center of this region. Extensive experiments using this reaction have not yielded any evidence for SHEs.^{3,6}

We have briefly discussed the importance of choosing a reaction capable of producing a SHE with a sufficiently long half-life and a large enough production cross section to be detectable. To enhance the survivability of fusion products requires a bombardment energy as low as possible while still permitting fusion. Some "extra-push" energy may also be necessary for fusion^{4,7} for this and several other fusion reactions capable of producing SHE's with ^{48}Ca projectiles. For these reasons, we chose two bombardment energies: one just at the estimated fusion barrier and one several MeV above to compensate for any "extra-push" energy that might be required for fusion in such a heavy system. The resultant excitation energies in the compound nucleus were 26 and 34 MeV, respectively.

We bombarded a target containing 1.14×10^{16} atoms/cm² ^{254}Es with ^{48}Ca at the SuperHILAC accelerator. At this time the target also contained 0.7×10^{16} atoms/cm² of ^{250}Cf from decay of the Es. We performed four separate bombardments, each lasting several hours, for a total of 7.7×10^{15} ^{48}Ca particles. The ^{48}Ca bombarding energies were 308 and 317 MeV, as measured with a surface-barrier detector. The calculated laboratory energies entering the Es deposit were then 240 MeV (the calculated laboratory fusion barrier) and 249 MeV, respectively, using the energy losses calculated from Hubert et al.⁸ The Es was electroplated in a 3.0 mm-diam spot on a 2.2 mg/cm² Be foil that had been coated with a 0.2 mg/cm² Pd layer to prevent attack of the Be by the electroplating solution. A 0.02 mg/cm² Pd layer was then vaporized over the Es oxide deposit to reduce transfer of Es to the recoil

collecting foils during bombardment. The foil arrangement in the target chamber is shown in Fig. 1.

Recoils from the Es target were collected on either beryllium or copper foils that were chemically processed to yield SHE fractions mounted on thin foils for coincident SF counting. Based on the predicted properties of SHEs,⁹⁻¹¹ both solution and gas-phase separation chemistries were performed. We designed these separations to recover elements 110 through 118. The primary fusion product, element 119, is predicted to decay rapidly^{1,2} by alpha emission into this range of elements and, thus, would be detected by the SF or, possibly, α -decay of its descendants.

Chemical yields of the naturally-occurring elements Pt through Po were used as substitutes for elements 110 (eka-Pt) through element 118 (eka-Rn). An aqueous separation procedure using sulfide precipitations and cation-exchange chromatography was performed during the one hour from the end of bombardment (EOB) to the beginning of counting. The counting samples were prepared by transferring the final sulfide precipitate to a $45 \mu\text{g}/\text{cm}^2$ carbon foil using a mixture of water and ethyl alcohol and subsequently evaporating the solution. The final sample contained about $80 \mu\text{g}$ of Au, Bi, and Pb carriers in the sulfide form. The recoil products from two of the bombardments, one at each energy, were processed using this method. Chemical yields were fairly consistent in bombardments A and B, except for Pb, as shown in Table I. The yield of Pb was apparently low because of slow elution from the cation-exchange column. Because eka-Pb is predicted to be more strongly complexed in HCl, its chemical yield should have been higher.¹²

Gas-phase and a subsequent aqueous separation were performed on the recoil products from the remaining two bombardments. This separation is based on the predicted increase in volatility and noble character of elements 112 through 116 in their elemental states in comparison to their naturally-occurring homologs.¹⁰ The copper recoil catcher foil was heated to 1050°C in a quartz apparatus using H_2 carrier gas. During the heating procedure the noble gases were condensed in a liquid nitrogen cooled cryogenic chamber and counted in situ for SF and α -decay within 30 min after EOB. Two fractions were condensed on Pd-covered Ni foils. These fractions included elements that condense in the range of $+5^\circ\text{C}$ to 400°C and

400°C to 1000°C, representing Hg and Pb-like SHEs, respectively. Counting for SF and α -decay in these fractions was begun 2 to 2 1/2 hours after EOB. After collecting these volatile fractions, we dissolved the copper foil and, finally, an aqueous SHE fraction was obtained from separations based on the predicted bromide complexation of SHEs.¹² Spontaneous-fission counting was begun on this fraction about four hours after EOB. Chemical yields for these fractions are presented in Table I (bombardments C and D).

Spontaneous-fission counting was performed in a fragment-fragment coincidence mode with opposing surface-barrier detectors. The counting efficiency for coincident fission events was about 70%, except for the noble gas (eka-Rn) fraction, which had a counting efficiency of 40% because of the use of a single annular surface-barrier detector. In more than two months of counting, no SF coincident events with total energies greater than 40 MeV were observed in any of the counting samples. A single event with 74-MeV energy was observed after 37 days in the sulfide fraction from the 240 MeV bombardment. A total of five low-energy (<40-MeV) coincident events were observed in the eka-Hg and eka-Pb fractions from the gas-phase separations. These events occurred between 10 h and 4 d after EOB with total kinetic energies of 22 to 36 MeV. The low energies and lack of later events led us to conclude that these events may have been caused by electronic noise, although we observed no such events in any of the calibrations with ²⁵²Cf or an alpha source. Measurements for some At and Pb isotopes were made in the SHE fractions from the gas-phase separations. Production cross sections for these isotopes (Table II) show an expected increase with energy above the fusion barrier.

Cross-section limits for SHEs were calculated on the basis of three events. This is the 95% confidence level for the observation of zero events.¹³ We present two curves in Fig. 2 for the higher-energy bombardments (B and D in Table I). Curve (a) presents the results for the experiments based on sulfide precipitations that collected all of the SHEs (except elements 117 and 118 and possibly eka-Pb) with similar chemical yields at the same time after EOB. Curve (b) shows cross-section limits for the eka-Pb and eka-Hg fractions from the gas-phase separations. The eka-Rn fraction would have lower cross-section limits for shorter half-lives because of faster processing of the sample, but would have considerably higher limits at late times because of poor chemical yield. Similarly, the SHE fraction obtained with bromide

complexation would have higher cross-section limits for shorter half-lives because of longer preparation times, but for longer half-lives the limits would be similar to the eka-Hg and eka-Pb curves in Fig. 2. We think the eka-Hg and eka-Pb fractions are more representative because most theoretical estimates predict that the longer-lived SHEs will be in these fractions. The lowest cross-section limit for SHE production is $2.5 \times 10^{-31} \text{ cm}^2$ for half-lives of a few days from these experiments. If both types of separations successfully collected SHEs, the lowest limit would be $1.5 \times 10^{-31} \text{ cm}^2$. The limits for the bombardments performed at the fusion barrier (A and C in Table I) would be nearly identical except for slightly higher limits at the shorter half-lives.

We believe that the $^{48}\text{Ca} + ^{254}\text{Es}$ reaction deserves further attention as a pathway to SHEs, particularly in view of the failure of the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction to produce SHEs. The LEAP¹⁴ proposal for the production of 40 μg of ^{254}Es would make possible a target 100 times larger than that used in our experiments. With a larger target and increased bombardment times, cross sections of nearly 10^{-35} cm^2 should be achievable.

It is also important to increase the sensitivity for short lifetimes. Randrup's calculations suggest that we cannot expect a long-lived SHE to result from alpha and beta decay of element 119, the primary fusion product. Instead, one or two alpha decays, several tens of milliseconds apart, would be followed by SF decay. These millisecond half-lives are accessible by other experimental techniques such as the velocity filter SHIP¹⁵ or a fast He-jet system.

This work was performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

References

- ¹J. Randrup, S. E. Larsson, P. Möller, A. Sobiczewski, and A. Lukasiak, *Physica Scripta* 10A, 60 (1974).
- ²E. O. Fiset and J. R. Nix, *Nucl. Phys.* A193, 647 (1972).
- ³P. Armbruster, *et al.*, *Phys. Rev. Lett.* 54, 406 (1985).
- ⁴S. Björnholm and W. J. Swiatecki, *Nucl. Phys.* A391, 471 (1982).
- ⁵G. A. Leander, P. Möller, J. R. Nix, and W. M. Howard, in *Proceedings of the Seventh International Conference on Atomic Masses and Fundamental Constants, Darmstadt, Seeheim, 1984*, edited by O. Klepper, (GSI, Darmstadt 1984) p. 466; P. Möller, private communication, 1985.
- ⁶E. K. Hulet *et al.*, *Phys. Rev. Lett.* 39, 385 (1977); J. D. Illige, *et al.*, *Phys. Lett.* 78B, 209 (1978); R. J. Otto *et al.*, *Inorg. Nucl. Chem.* 40, 589 (1978); A. Ghiorso *et al.*, Lawrence Berkeley Laboratory Report No. LBL 6575, 1977 (unpublished); Yu. Ts. Oganessian *et al.*, *Nucl. Phys.* A294, 213 (1978).
- ⁷H. Gäggeler, T. Sikkeland, G. Wirth, W. Bröchle, W. Bögl, G. Franz, G. Herrmann, J. V. Kratz, M. Schädel, K. Sümmerer, and W. Weber, *Z. Physik* A316, 291 (1984).
- ⁸F. Hubert, A. Fleury, R. Bimbot, and D. Gardes, *Ann. Phys., Fr.* 5 (Suppl) (1980).
- ⁹B. Fricke, *Structure and Bonding* 21, 89 (1975).
- ¹⁰B. Eicher, *Kernenergie* 19, 307 (1976).
- ¹¹K. S. Pitzer, *J. Chem. Phys.* 63, 1032 (1975).
- ¹²J. V. Kratz, J. O. Liljenzin, and G. T. Seaborg, *Inorg Nucl. Chem. Lett.* 10, 951 (1974).
- ¹³D. B. Owen, *Handbook of Statistical Tables* (Addison-Wesley, Reading, Ma., 1962).

¹⁴A. Ghiorso, D. Hoffman, E. K. Hulet, O. L. Keller and G. T.

Seaborg, LEAP (Large Einsteinium Accelerator Program), Lawrence Berkeley Laboratory

PUB-5118 (1984), unpublished.

¹⁵G. Münzenberg et al., Nucl. Instrum. Methods 161, 65 (1979).

Figure captions and titles:

Fig. 1. A schematic diagram of the foil arrangement in the target chamber. (a) 1.8 mg/cm² HAVAR window. HAVAR is a trademark of the Hamilton Precision Metal company, Lancaster, Pa., and contains mostly Co, Cr, Ni, and Fe. (b) Ta collimator. (c) The Es target electroplated on a Be foil. Nitrogen gas (0.2 mg/cm²) is directed against the HAVAR and Be for cooling. (d) A 30. µg/cm² Al foil to prevent transfer of the target isotope to the recoil foil. (e) A 6.3 mg/cm² Cu or 2.2 mg/cm² Be recoil-catcher foil. The 17.4 mm-diam catcher foil was 5 mm from the target.

Fig. 2. Cross-section limits for the production of SHEs as a function of half-life for the two bombardments at 249 MeV. (a) Cross-section limits from the sulfide precipitations of bombardment B. A chemical yield of 55% was used to calculate the limits. (b) Cross-section limits for eka-Pb and eka-Hg from the gas-phase separations of bombardment D.

Table I. Bombardment parameters and chemical yields for SHE homologs. Some yields were determined by the addition of tracers prior to the chemical separations for these bombardments. Others were estimated based on separate experiments.

<u>Bombardment</u>		$^{48}\text{Ca}^{14+}$ energy enter- ing the Es (MeV)	<u>Chemical yield (percent)</u>									
<u>Time</u> <u>(min)</u>	<u>Fluence</u> <u>($\times 10^{-15}$)</u>		Z: eka:	<u>110</u> <u>Pt</u>	<u>111</u> <u>Au</u>	<u>112</u> <u>Hg</u>	<u>113</u> <u>Tl</u>	<u>114</u> <u>Pb</u>	<u>115</u> <u>Lp</u>	<u>116</u> <u>Po</u>	<u>117</u> <u>At</u>	<u>118</u> <u>Rn</u>
A 773	2.22	240		40	44	48	47	18	54	43	a	a
B 383	2.14	249		47	53	55	53	21	61	69	a	a
C 338	2.02	240		85	85	75	85	80	85	85	85	10
D 393	2.19	249		85	85	75	85	80	85	85	58	10

^aNot collected.

Table II. Cross sections for At and Pb isotopes from the gas-phase separations.

<u>Bombardment energy (MeV)</u>	<u>Isotope</u>	<u>Cross Section (nb)</u>
240	^{211}At	$290. \pm 120.$
240	^{212}Pb	$140. \pm 30.$
249	^{209}At	$1500. \pm 400.$
249	^{211}At	$1100. \pm 200.$
249	^{212}Pb	$590. \pm 120.$

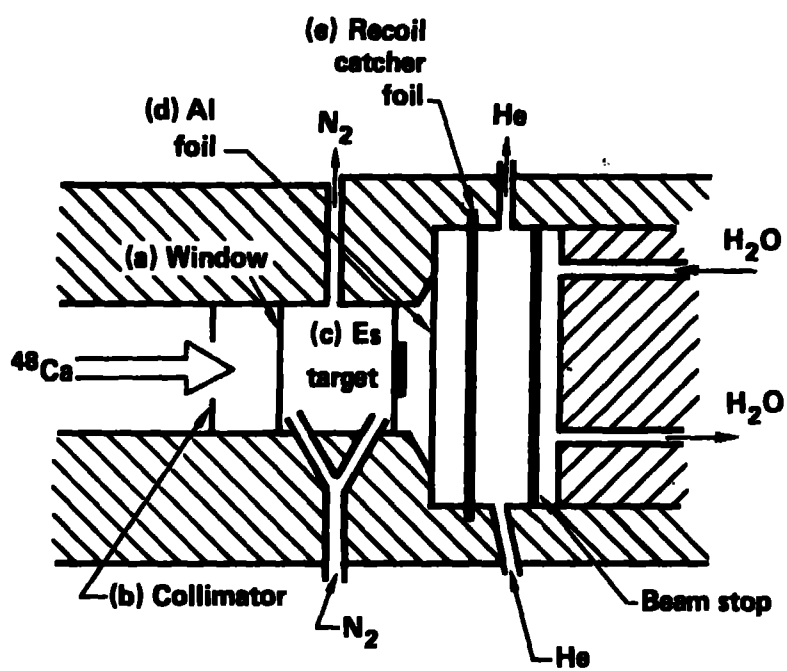


Figure 1

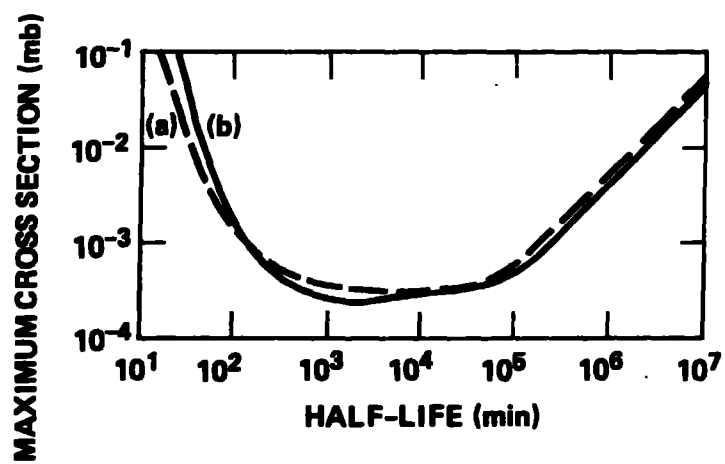


Figure 2